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Tunable Lasers and Coherent Light Techniques

for High Resolution Ultraviolet Spectroscopy

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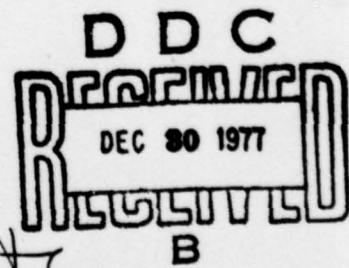
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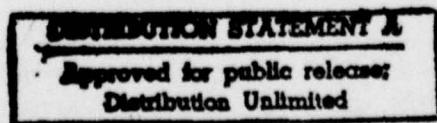
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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

This report summarizes recent efforts towards the development, improvement, and use of tunable laser systems and novel coherent light techniques for high resolution ultraviolet spectroscopy of atoms and molecules. It reports on: (a) studies of atomic hydrogen and deuterium by Doppler-free ultraviolet laser spectroscopy, which have yielded new precision values of the 1S-2S isotope shift and of the 1S ground state Lamb shift; (b) the first demonstration of a novel technique of high resolution laser spectroscopy, using coherent two-photon excitation with multiple light pulses: (c) the status of a search for a

over >

19. Polarization spectroscopy
Lamb shift of 1S
1S-2S Isotope shift
Multi-Pulse Two-Photon Excitation
Ramsey Fringes

20. hydrogen-laser-pumped tunable ultraviolet laser; *and* (d) the observation of a quenching of the laser action in xenon fluoride excimer lasers by the vapor of xenon difluoride. This report contains 4 reprints:

T.W. Hänsch, "New Methods of Laser Spectroscopy"
C. Wieman and T.W. Hänsch, "Precision Measurement of the Ground State Lamb Shift in Hydrogen and Deuterium"
R. Teets, J. Eckstein, and T.W. Hänsch, "Coherent Two-Photon Excitation by Multiple Light Pulses"
T.W. Hänsch, "Multiple Coherent Interactions".

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CONTENTS

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A. HIGH RESOLUTION SPECTROSCOPY OF ATOMIC HYDROGEN AND DEUTERIUM

Extending earlier experiments (1,2) the 1S-2S transition of atomic hydrogen and deuterium was studied by Doppler-free two-photon spectroscopy. The required intense monochromatic ultraviolet radiation near 243 nm was generated with a unique laser system, developed under ONR support, and consisting of a cw dye laser oscillator (λ=486 nm), a nitrogen-pumped pulsed dye laser amplifier and a lithium formate frequency doubler. Thanks to the near Fourier-transform limited line width of this ultraviolet laser system, the isotope shift of the 1S-2S transition could be measured to within about 1 part in 100 000. The measurement confirms the existence of the theoretically predicted 24 MHz "Dirac Shift" of the hydrogen ground state due to relativistic nuclear recoil effects. An improvement in accuracy by another order of magnitude will yield a new precision value of the electron/proton mass ratio

The blue Balmer beta line (n=2-4) has been simultaneously studied by Doppler-free polarization spectroscopy, using the fundamental cw dye laser output. The Balmer beta interval is almost exactly equal to one quarter of the 1S-2S interval, and the two intervals can be compared very precisely, because the two resonances are observed at almost the same laser frequency. These measurements have yielded a new experimental value of the hydrogen 1S ground state Lamb shift with an accuracy better than 4 parts in 1000, well exceeding the accuracy of an earlier comparison, using a saturated absorption spectrum (2). The main source of uncertainty has been a small frequency shift and chirping, introduced by the pulsed amplifies. A preliminary description of

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these experiments can be found in two papers, "New Methods of Laser
Spectroscopy" by T. W. Hänsch, and "Precision Measurement of the Ground State
Lamb Shift in Hydrogen and Deuterium" by C. Wieman and T. W. Hänsch, which are
included as Appendix A and Appendix B respectively. A more detailed account is
under preparation.

B. DOPPLER-FREE TWO-PHOTON SPECTROSCOPY WITH MULTIPLE COHERENT LIGHT PULSES

Doppler-free two-photon excitation of atoms has been observed with a train of
phase-coherent standing wave light pulses. In initial experiments, a pulse train
was produced by multiple reflections of a single laser pulse inside an optical
resonator, and the 3s-5s transition of atomic sodium was studied (3). Quantum
interference effect result in narrow spectral lines, similar to Ramsey fringes,
whose width depends only on the resonator losses and the natural transition line
width, not on the laser line width. The resonant excitation probability is
proportional to the square of the number of light pulses incident during the
atomic relaxation time, and a dramatic signal enhancement over single pulse
excitation is possible. We are presently exploring the feasibility of using a
mode-locked cw dye laser for such experiments.

Two-photon excitation with multiple light pulses promises to extend the range of
high resolution Doppler-free laser spectroscopy to new wavelength regions, in
particular in the ultraviolet and vacuum ultraviolet, where only short pulse
laser sources are available at present. The principles and early experimental
results are discussed in two papers "Coherent Two-Photon Excitation by Multiple

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Light Pulses" by R. Teets, J. Eckstein, and T. W. Hänsch, and "Multiple Coherent Interactions" by T. W. Hänsch, which are included as Appendix C and Appendix D.

C. TUNABLE ULTRAVIOLET LASERS

C1. Search for Hydrogen-Laser-Pumped UV Laser

Although tunable coherent ultraviolet radiation can be generated by frequency mixing in nonlinear optical materials, this approach tends to be relatively inefficient and complex. A desirable alternative would be a system similar to a nitrogen-laser-pumped dye laser but with suitable substitutes for pump and active material to permit direct laser operation at shorter wavelengths. This approach has already provided some success with the use of a KrF laser to pump the dye p-terphenyl, producing tunable laser output over the range 335-346 nm (4).

We have attempted to reach considerably shorter wavelengths by using a vacuum ultraviolet hydrogen laser (5) as a pump source. The laser produces 40 kW pulses of approximately .5 nsec duration at λ=161 nm and is very similar in its properties to a nitrogen laser, including simplicity of construction and low operating costs. Unfortunately it has proven very difficult to find a material that can be effectively pumped at this wavelength. Many simple hydrocarbon compounds have absorption bands near 161 nm, but their fluorescence quantum efficiency is generally quite low. Some non-organic materials, in particular rare-earth doped trifluorides (6), looked considerably more promising.

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We have obtained a sample of $LiYF_4$:Pr (.25 At.%) from Dr. A. Linz of the MIT
Crystal Physics Laboratory. This material has an observed fluorscence quantum
yield of about 80% when excited by radiation at λ= 160 nm and a predicted
"tuning range" of 215 - 260 nm. A 3 mm long polished crystal was placed in a
simple 4 cm long optical cavity and a thin region near the surface was
transversely pumped by the VUV laser light, focused to a narrow line.
Unfortunately we were unable to produce laser action, despite strong ultraviolet
fluorescence. The gain was obviously insufficient to overcome the losses in the
crystal, augmented by excited state absorption. The development of a
hydrogen-laser-pumped tunable ultraviolet laser system depends still on the
discovery of some other more suitable laser material.

C2. Xenon Difluoride in Xenon Fluoride Excimer Lasers

Discharge pumped xenon fluoride excimer lasers appear as a very attractive
alternative to nitrogen lasers for the pumping of pulsed dye laser systems.
They not only offer greater efficiency, but also considerably longer pulse
length which is particularly important for the generation of highly
monochromatic laser radiation. But although commercial versions of such lasers
are becoming available from several manufacturers, they still suffer from a
serious problem which precludes any economical and simple long-term operation:
the rather expensive gas mixture of helium, xenon, and some fluorine donor
(typically a NF_3 or F_2 at a concentration of a few parts in 1000) has to be
replaced at short intervals, because the very active fluorine is consumed by

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various chemical reactions.

We have attempted to solve this problem by finding a suitable solid or liquid

fluorine donor with proper vapor pressure which would automatically maintain a

constant fluorine concentration. Xenon difluoride (7), a stable white powder,

appeared as a particularly promising candidate. This compound has a vapor

pressure of about 4 Torr at room temperature, and its dissociation should not

introduce any unwanted foreign substances into the gas discharge. We have

obtained some samples of highly pure XeF_2 from Prof. N. Bartlett at the

Chemistry Department of the University of California at Berkeley. The vapor of

this substance was introduced into the He/Xe or $He/Xe/NF_3$ mixture inside two

different Blumlein-type transverse discharge configurations, including a

well-tested excimer laser of Dr. Charles Wang at Aerospace Corporation in Los

Angeles. The vapor pressure was varied from 4 to 10 torr by slightly heating

the reservoir and discharge channel. Unfortunately, no laser action could be

obtained over a wide range of operating conditions, and the introduction of a

few Torr of XeF_2 vapor into an operating XeF laser actually quenched any laser

action. The mechanism of this quenching is not yet fully understood, but these

experiments may nonetheless point the way for improved gas handling in XeF

lasers: since XeF can form spontaneously in the discharge of a XeF excimer

laser, the removal of this substance would seem important to prolong the

operating time and increase the power of such lasers.

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D. REFERENCES

(1) T. W. Hänsch, S. A. Lee, R. Wallenstein, and C. Wieman, Phys. Rev. Letters 34, 307 (1975)

(2) S. A. Lee, R. Wallenstein, and T. W. Hänsch, Phys. Rev. Letters 35, 1262 (1975)

(3) R. Teets, J. Eckstein, and T. W. Hänsch, Phys. Rev. Letters 38, 760 (1977)

(4) D. G. Sutton, G. A. Capelle, Appl. Phys. Letters 29, 563 (1976)

(5) J. E. M. Goldsmith and I. N. Knyazev, J. Appl. Phys. (to be published)

(6) K. H. Yang and J. A. DeLuca, Appl. Phys. Letters 29, 499 (1976)

(7) J. H. Holloway, "Noble Gas Chemistry," London, 1968

NEW METHODS OF LASER SPECTROSCOPY[*]

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New techniques of nonlinear laser spectroscopy provide powerful
tools for studies of atoms and molecules: *Coherent two-photon
excitation with multiple light pulses* produces narrowband
spectral interference fringes and strongly enhanced resonant
signals, and holds promise for very high resolution spectroscopy
of atomic hydrogen. *Laser polarization spectroscopy* is a method
of Doppler-free spectroscopy, which greatly surpasses saturated
absorption spectroscopy in sensitivity, by monitoring the
nonlinear interaction of two laser beams in a gas via changes in
light polarization rather than intensity. The method has been
very successfully used for a study of the hydrogen Balmer β line.
Polarization labeling with laser light can unravel the complexities
of molecular absorption spectra by identifying all absorption lines
with a common lower level.

INTRODUCTION

Laser techniques have been particularly successful in overcoming the Doppler
broadening of spectral lines, which is caused by thermal motion and often blurs
important details in the line structure and line shape of free atoms or molecules.
Rather spectacular resolutions can be obtained by saturated absorption spectroscopy
[1] and by Doppler-free two-photon spectroscopy [2] of gas samples, without any need
to resort to the classical scheme of transverse observation of a well collimated
molecular beam. This paper will report on some interesting advances in these
techniques, which have been successfully demonstrated during the past year, and
which can greatly enhance the sensitivity, resolution and range of application of
high resolution laser spectroscopy.

TWO-PHOTON SPECTROSCOPY WITH MULTIPLE LIGHT FIELDS

A particularly simple and elegant technique of laser spectroscopy without Doppler
broadening is the method of two-photon excitation with two counterpropagating
monochromatic laser beams, whose first order Doppler shifts cancel. This scheme,
which was first experimentally demonstrated about three years ago, has already been
used widely for novel studies of atoms and molecules [2]. Typically, a gas sample
is placed in a standing wave laser field, and the excitation is monitored by
observing the fluorescence of the excited atoms. All the atoms contribute to the
narrow Doppler-free resonance signal, not just a few with selected velocities. But
the excitation probability is proportional to the square of the laser intensity, and
the laser source should be both intense and highly monochromatic, two obviously
contradicting requirements, in particular, if frequency mixing in nonlinear optical
crystals is necessary to generate the required wavelength.

This problem has been the main obstacle in some recent studies of the two-photon
transition from the 2S ground state of atomic hydrogen to the metastable 2S state,
carried out at Stanford [3,4]. The upper state has a natural lifetime of 1/7 sec and
should in principle permit an unprecedented resolution of better than 1 part in 10^{15}.
Precision spectroscopy of this simplest of the stable atoms is of obvious interest
because it can provide accurate values of fundamental constants and provide stringent

tests of quantum electrodynamic calculations.

In the quoted experiments, the required ultraviolet radiation at 2430Å was generated by frequncy doubling the output of a powerful pulsed dye laser oscillator amplifier system. But the short pulse length of about 10 nsec limited the resolution to line widths not better than about 60 MHz. Nonetheless, Carl Wieman in our laboratory has been able to measure the 1S-2S isotope shift for hydrogen and deuterium to within about 5 MHz or 1 part in 10^5 in this way, and he has, for the first time, obtained experimental evidence for the existence of a theoretically predicted 24 MHz "Dirac-shift" of the hydrogen 1S ground state, associated with the nuclear recoil motion in this relativistic two-body system.

Another factor of twenty improvement in accuracy would provide a new value for the electron/proton mass ratio, and an absolute wavelength measurement to that accuracy would determine a new Rydberg constant with a hundredfold improvement in precision over the best previous measurement [5].

The most obvious way to overcome the Fourier-transform limits of pulsed laser spectroscopy is the use of a cw laser. Highly monochromatic cw radiation at 2430Å has indeed been generated at Stanford by summing the frequency of a blue krypton ion laser and a yellow dye laser in cooled ADP. But the low power of only about .1 mW would require tight focusing for two-photon excitation of hydrogen 1S-2S, and then the short transit time of the atoms, moving through the narrow beam waist, would again broaden the observed line width.

A rather ingenious solution to this transit time problem has been suggested by Baklanov et al. [6]. An atomic beam can be sent through two consecutive transverse standing light fields, separated in space. After passing through the first field, the atoms are found in a coherent superposition of states and oscillate at the two-photon resonance frequency. The effect of the second light field depends on the phase of the radiation relative to the atomic oscillations, so that atoms passing through this field will be either further excited or returned to the ground state by stimulated two-photon emission. When monitoring the net excitation by the two fields one should observe the optical analog of the well known Ramsey fringes, which are routinely used in rf spectroscopy of atomic beams [7]. The spectral resolution is then limited by the travel time between the two fields rather than by the transit time through each waist. Doppler-free two-photon excitation with standing waves ensures that the fringe structure is not smeared out by dephasing due to the unavoidable small spread of transverse atomic velocities.

Attempts to use these narrow spectral fringes for very-high-resolution spectroscopy of atomic beams are presently under preparation in several laboratories. Similar fringes can also be observed by saturated absorption spectroscopy with spatially separated laser fields, as recently demonstrated by Bergquist et al. [8] for a fast beam of atomic neon.

To an atom traversing the separated field regions, the laser field appears as a succession of light pulses. This suggests that optical Ramsey fringes in two-photon excitation are also observable with a pulsed laser source, without any need for an atomic beam, if a gas cell is irradiated by two standing wave light pulses, separated by a delay time T. If the pulse delay time T is scanned together with the wavelength, the spacing between adjacent fringes will be equal to 1/T. Such interference fringes have indeed been observed by Salour and Cohen-Tannoudji [9] for the sodium 3S-4D transition. The observation of spectral fringes of a width well below the Fourier-transform limit of an individual laser pulse opens new prospects for high resolution spectroscopy, although the sinusoidal fringe structure would make it rather difficult to resolve spectral components that are very closely spaced.

These excitation experiments using two separated fields can be regarded as analogs to optical diffraction at a double slit. This analogy suggests an important next step: it is also possible to observe the analog to diffraction at a multiple slit array or diffraction grating. If two-photon transitions are excited by a whole train of

phase-coherent light pulses, the spectral fringes condense into narrow lines which can be compared to the diffraction orders of a grating or to the multi-beam interference fringes of a Fabry-Perot interferometer.

Such narrow multi-pulse interference fringes have recently been observed at Stanford by Teets et al. [10] for the 3S-5S transition of atomic sodium with a very simple experimental setup. A pulse train is produced by injecting a single short dye laser pulse into an optical cavity, formed by two mirrors. The gas sample is placed near one end mirror so that the atoms see a pulsed standing wave field once during each roundtrip, when the pulse is being reflected by the mirror. For spectral fine tuning, the resonator length is scanned with a piezotranslator. The observed sharp interference fringes can be interpreted as originating from the "shock-excited" narrowband modes of the optical resonator.

The signal magnitude can be easily predicted by analyzing the excitation in the time domain, using perturbation theory. A dramatic signal increase, compared to single-pulse excitation, is expected if the laser pulse is injected into the resonator without loss, for instance by a fast electro-optic light valve. As long as atomic relaxation can be neglected, the probability of two-photon excitation for small intensities is proportional to the square of the number of pulse roundtrips.

Two-photon excitation with multiple light pulses promises to extend the range of Doppler-free laser spectroscopy to new wavelength regions, in particular in the ultraviolet and vacuum ultraviolet, where only short pulse laser sources are available at present. The technique may make it quite feasible to improve the resolution of the 1S-2S transition of hydrogen to about 1 part in 10^{10}, as limited by the relativistic transverse Doppler effect at room temperature.

HIGH RESOLUTION POLARIZATION SPECTROSCOPY

For normal single-photon transitions, Doppler-broadening can only be reduced by restricting the range of atomic velocities along the direction of observation. In the saturation techniques [1], intense monochromatic laser light is used to "label" atoms in a narrow range of axial velocities by selectively removing them from an absorbing level. A second laser beam probes the resulting bleaching of the gas sample and records a Doppler-free saturation spectrum.

At Stanford, Wieman [12] has demonstrated a new technique of Doppler-free spectroscopy, which is related to saturation spectroscopy, but can provide considerably higher sensitivity and signal-to-noise ratio than the older approach. In this method of "polarization spectroscopy," atoms of a gas sample in a selected velocity range are labeled by orienting them through optical pumping with circularly polarized laser light. These labeled atoms can then be detected with high sensitivity by observing their effect on the polarization of a probe laser beam.

A polarization spectrometer can be rather simple. The gas sample is contained in a cell with good, strain-free optical windows. The output of a monochromatic tunable laser is divided into a circularly polarized saturating beam and a linearly polarized probe beam, which are sent in opposite directions through the same sample region. A crossed polarizer prevents probe light from reaching a detector, as long as the sample is optically isotropic.

Normally, in a gas, atoms have their rotation axes distributed at random in all directions. The polarized laser beam preferentially removes those with particular orientations, leaving the remaining atoms polarized. The linearly polarized probe light can be thought of as a combination of right-handed and left-handed circularly polarized waves of equal intensity. If the laser is tuned near the center of a Doppler-broadened line, where the probe beam can "see" the atoms polarized by the pump beam, those with zero axial velocity, more of the left-handed wave, say, is absorbed than of the right-handed one, and the light passing through the sample becomes elliptically polarized. In addition, the polarization axis is rotated,

because the two waves also see different refractive indices. The probe beam thus acquires a component that can pass through the crossed polarizer in front of the detector.

Polarization spectroscopy has an important advantage over saturated absorption spectroscopy, because there is almost no transmission of the probe beam until its polarization is changed by the pumped atoms. With essentially no background, the signal is not easily obscured by noise or intensity fluctuations, and the method can be used with fewer atoms and lower laser power.

For very small signals it is actually advantageous to uncross the polarizers slightly. The detector will then register some finite background, but the polarization signal appears enhanced by its interference with this background. The resulting resonance curve is dispersion shaped, and facilitates, for instance, electronic locking of the laser frequency to an absorption line.

This technique of polarization spectroscopy was developed in response to the need for a more sensitive method of Doppler-free "single-photon" spectroscopy which had become painfully apparent during a study of the blue Balmer-β line of atomic hydrogen at 4960Å by saturated absorption spectroscopy [4]. The Balmer-β energy interval (n=2-4) is almost exactly equal to one quarter of the Lyman-α interval (n=1-2), and the same dye laser, whose second harmonic is used to study the 1S-2S two-photon transition, can simultaneously record the Balmer-β line at its fundamental wavelength. The two optical intervals can be compared very precisely in this way, and from such a comparison, Lee et al. [4] were able to determine, for the first time, the Lamb shift of the 1S ground state of hydrogen to an accuracy of almost 1%. The main limitation was the relatively poor resolution of the Balmer-β saturation spectrum. This line is considerably more difficult to observe than the red Balmer-α line [13], because the fine structure splitting of the upper level is smaller. In addition, this line is more susceptible to splitting by the electric field in the discharge tube, used to form the atomic hydrogen. It is also much weaker and so requires a stronger discharge and a more intense saturating laser beam. Because of all these factors, it was not possible to resolve single fine structure components by the old saturated absorption method.

The polarization spectra, obtained from the same hydrogen Balmer-β line with a cw dye laser, give dramatic evidence for the value of the technique, showing strikingly clean signals with relatively few atoms (1-2% absorption) and linewidths as narrow as 25 MHz. These spectra did not only resolve single fine structure components, but revealed, for the first time, the Stark shifts arising from the weak (10V/cm) electric field in the glow discharge. The data from a new precision measurement of the ground state Lamb shift, using such polarization spectra as reference, are presently being evaluated by Carl Wieman at Stanford as part of his thesis research and promise an at least tenfold improvement in accuracy.

SIMPLIFICATION OF SPECTRA BY POLARIZATION LABELING

Polarization spectroscopy is not only a powerful tool to study fine spectral details with high resolution, it can also be used to unravel and greatly simplify complex molecular absorption spectra, as demonstrated by Teets et al. at Stanford [12] in experiments with diatomic sodium molecules.

Ordinarily, very many of the rotational-vibrational levels of the electronic ground state of a molecule are populated, and the optical absorption spectrum is a complicated superposition of absorptions from each of these ground state levels. But the polarization method can be used to label and chosen level by orienting its molecules with a circularly polarized laser beam, tuned to a selected absorption line. Alternatively, the molecules can be aligned by pumping with linearly polarized light. All other lines, originating from the same lower level, can then be easily identified, because the sample will change the polarization of a probe beam at these wavelengths. Labeling of molecular levels by depleting selected levels through saturation and

monitoring the resulting reduced absorption had been demonstrated before by Kaminsky et al. [13], but polarization spectroscopy offers again a considerably higher sensitivity.

To quickly survey a large spectral region, one can use a broadband probe beam, as generated by a dye laser without wavelength selective tuning elements. This probe light is sent through the sample between crossed polarizers and into a spectrograph. The lines originating from the labeled lower level appear as a simple series of bright lines on a dark background and resemble the spectra of laser-excited fluorescence, but give direct information about the quantum numbers and parameters of the electronically excited upper state rather than the ground state.

Such spectra are immensely helpful for unraveling complex spectra and for studying perturbations and collisions. Once a spectral line is identified, it can be studied with high resolution, free of Doppler broadening, and its wavelength can be measured with high precision. With accurate fringe counting digital wave meters [14] it is thus easily possible to determine much improved spectroscopic parameters, even for molecules that were as thoroughly studied in the past as diatomic sodium.

CONCLUSION

We have only been able to look at a few new methods of laser spectroscopy, which have been developed and explored at Stanford during the past year or so. Numerous other powerful spectroscopic techniques using lasers had to remain unmentioned. But the few examples may be sufficient to convince us that we can expect a rich harvest of important results - and perhaps surprising discoveries - from these and other new laser techniques in the future.

REFERENCES

1 V. S. Letokhov, in *High Resolution Laser Spectroscopy*, K. Shimoda, ed., Springer, New York (1976), p 95.

2 N. Bloembergen and M. D. Levenson, ibid., p 315.

3 T. W. Hänsch, S. A. Lee, R. Wallenstein, and C. Wieman, Phys. Rev. Letters 34, 307 (1975).

4 S. A. Lee, R. Wallenstein, and T. W. Hänsch, Phys. Rev. Letters 35, 1262 (1975).

5 T. W. Hänsch, M. H. Nayfeh, S. A. Lee, S. M. Curry, and I. S. Shahin, Phys. Rev. Letters 32, 1396 (1974).

6 Ye. V. Baklanov, V. P. Chebotayev, and B. Ta. Dubetsky, Appl. Phys. 11, 201 (1976).

7 N. F. Ramsey, *Molecular Beams*, Oxford UP, London (1956), p 124.

8 J. C. Bergquist, S. A. Lee, and J. L. Hall, Phys. Rev. Letters 38, 159 (1977).

9 M. Salour and C. Cohen-Tannoudji, Phys. Rev. Letters 38, 757 (1977).

10 R. Teets, J. Eckstein, and T. W. Hänsch, Phys. Rev. Letters 38, 760 (1977).

11 C. Wieman and T. W. Hänsch, Phys. Rev. Letters 36, 1170 (1976).

12 R. Teets, R. Feinberg, T. W. Hänsch, and A. L. Schawlow, Phys. Rev. Letters 37, 683 (1976).

13   M. E. Kaminsky, R. T. Hawkins, F. V. Kowalski, and A. L. Schawlow, Phys. Rev. Letters 36, 671 (1976).

14   F. V. Kowalski, R. T. Hawkins, and A. L. Schawlow, J. Opt. Soc. Am. 66, 965 (1976.

APPENDIX B

PRECISION MEASUREMENT OF THE GROUND STATE LAMB SHIFT IN HYDROGEN AND DEUTERIUM*

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The measurement of the $2S_{1/2}$ state Lamb shift in hydrogen was one of the main stimuli for the development of quantum electrodynamics. Since the first experiment by W. E. LAMB and R. C. RETHERFORD [1] the Lamb shift has been measured for a great many hydrogenic states, and such measurements are among the most precise tests of QED [2]. One measurement which was conspicuously missing from this list, however, was the Lamb shift of the hydrogen ground state. This has only been measured in the past few years [3,4] and the experiment described here is a continuation of those measurements to higher precision by the use of several technical innovations. We have also made an improved measurement of the Lyman-α hydrogen-deuterium isotope shift sufficiently precise to give the first experimental confirmation of the predicted relativistic nuclear recoil correction. The technical improvements mentioned are the development of a highly sensitive new form of Doppler-free spectroscopy, polarization spectroscopy, and the use of a single frequency cw dye laser at 4860Å with a high power pulsed amplifier system.

The shift of the 1S state energy from the value predicted by simple Dirac theory, which we call the Lamb shift, is calculated to be 8149 MHz [5]. QED corrections are the dominant contribution, but the shift also includes some small nuclear effects. Although this is eight times larger than the shift of the 2S state it cannot be measured using the traditional RF approach because there is no nearby P state reference level. Also there are several difficulties in measuring the Lamb shift by determining the Lyman-α wavelength to sufficient precision using conventional absorption or emission spectroscopy. The most serious of these are the 40 GHz Doppler width at room temperature, and the difficulty in making precision wavelength measurements in the vacuum ultraviolet region (1215Å). Moreover, the present uncertainty in the Rydberg constant precludes any such Lamb shift measurement to better than one part in 10^3.

The recent development of several nonlinear laser spectroscopic techniques, and improved tunable dye lasers has made it possible to determine the 1S Lamb shift in quite a different manner, however. This approach, as first described in [3], uses laser spectroscopy to compare the Balmer-β (n=2 to n=4) transition energy with 1/4 the Lyman-α energy. The comparison is made using a high power tunable narrowband laser at 4860Å, whose frequency doubled output at 2430Å excites the two-photon transition from the 1S ground state to the metastable 2S state. Simultaneously the Balmer-β line is observed using the fundamental laser output. If the Bohr formula were correct this interval would be exactly 1/4 the Lyman-α interval and we would find the two

resonances at exactly the same fundamental frequency. The actual displacement is due to the ground *state Lamb shift* plus small well measured relativistic and QED corrections to the excited state energies. So an accurate comparison of the two optical energy intervals allows one to measure the unknown 1S Lamb shift. This approach avoids the technical problems of VUV spectroscopy since one only measures visible wavelengths. In addition, the use of counterpropagating beams to excite the 1S-2S two-photon transtion eliminates the Doppler broadening, as discussed in [6]. Because this measurement involves only the comparison of two transitions in hydrogen the uncertainty in the Rydberg constant does not matter.

The first experimental value [3] for the 1S Lamb shift was obtained by exciting the 1S-2S transition just as described, and recording a simple Doppler-broadened laser absorption spectrum of the Balmer-β line as the reference. Substantial improvement was obtained by using the technique of saturated absorption spectroscopy with a pulsed laser to observe the Balmer-β line [4]. This gave sub-Doppler resolution, but the limitation of the quoted Lamb shift accuracy was still the poor resolution of the Balmer-β line.

The present experiment, which is shown schematically in Fig. 1, measured the Balmer-β line with enormously improved resolution by using a single frequency cw dye laser and the technique of polarization spectroscopy. The Balmer-β portion of the experiment has been previously described in [7], but will be briefly reviewed.
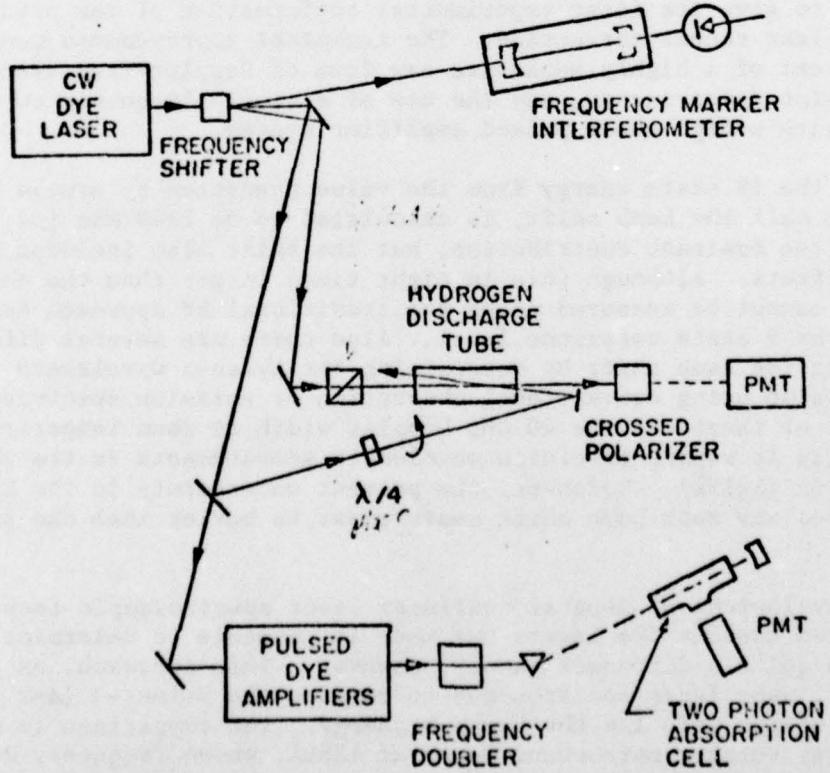

Fig. 1. Schematic of experimental apparatus

The laser was a cw jet stream dye laser (modified Spectra-Physics Model 375) using coumarin-one in ethylene glycol pumped by a UV argon laser (Spectra-Physics #171). Single frequency operation was achieved by having three intracavity etalons. At 1.5 W pump power the laser provides a single mode output of about 10 mW near 4860Å with a linewidth of ~ 3 MHz and continuously tunable over 4 GHz.

The technique of polarization spectroscopy uses induced optical anisotropy as a sensitive means to obtain Doppler-free line profiles. The basic schematic is shown in the middle portion of Fig. 1. A weak linearly polarized probe beam from the cw laser is sent through a Wood's type hydrogen discharge tube. Only a small fraction of this beam reaches a photodector after passing through a nearly crossed linear polarizer. Any optical anisotropy which changes the probe polarization will alter the light flux through the polarizer and can be detected with high sensitivity. Such an anisotropy can be induced by sending a second, stronger, circularly polarized beam from the laser in nearly the opposite direction through the discharge tube. Because the beams are counterpropagating their interactions are Doppler free. The improved resolution obtained using this technique enabled us to measure and correct for the effects of Stark and pressure shifts in the discharge.

Although the cw laser was highly advantageous in observing the Balmer-β line its power was much too low to excite the 1S-2S transition. Therefore, it was necessary to send the cw beam through a series of three pulsed dye amplifiers pumped by a 600 KW nitrogen laser (Molectron UV 1000). With a 1 mW input these gave output pulses 7 nanoseconds long with ~ 100 kW peak power and about 5% pulse to pulse amplitude fluctuation. The fundamental linewidth was about 120 MHz which gives two-photon linewidths less than one quarter as wide as those obtained with the previously used pulsed oscillatoramplifier system [4].
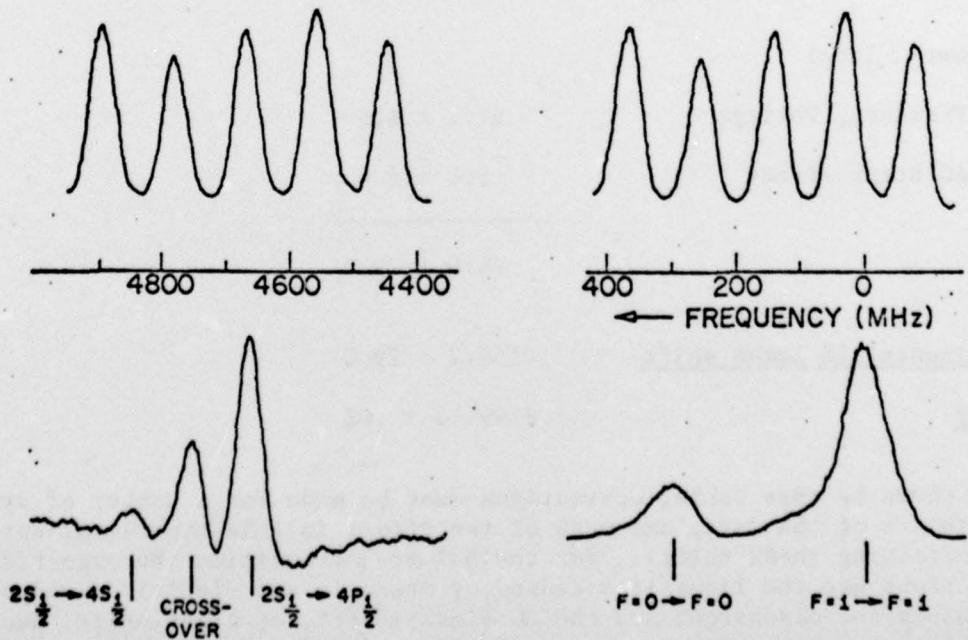

Fig. 2. Portion of the hydrogen Balmer-β polarization spectrum (left) and 1S-2S two-photon spectrum (right)

The two-photon spectrometer, as shown at the bottom of Fig. 1, is identical to that described in [3,4]. The output of the pulsed amplifier system goes into a lithium formate frequency doubling crystal which is angle tuned to phase match near 4860Å. The output of this crystal goes through a quartz prism which separates the doubled light from the fundamental. The doubled light passes through a hydrogen absorption cell, a small arm attached to the side of a Wood dicharge tube, and is reflected back on itself to obtain the Doppler-free 1S-2S excitation. This excitation is monitored by observing the collision induced 2P-1S fluorescence at 1215Å, through a MgF_2 window, with a photomultiplier.

In Fig. 2 we show typical spectra which were obtained. On the lower left is the portion of the Balmer-β transition used as a reference, while on the lower right is the 1S-2S two-photon spectrum. On the top of the figure are shown the frequency calibration lines obtained by sending a portion of the cw laser beam through the semiconfocal calibration interferometer shown at the top of Fig. 1.

The experimental results for hydrogen are shown in Table I.

TABLE I - Hydrogen 1S Lamb Shift

Uncorrected measurement: 8133.2 ± 4.8 MHz

Corrections:

  1S Two-Photon Spectrum:

  AC Stark Effect $3.2 \pm .8$ MHz

  Amplifier shift/line shape $- 40.0 \pm 28.0$

  Balmer-β line:

  Pressure, Voltage 57.2 ± 6.8

  AC Stark effect 5.6 ± 2.0
  _____
  Total 26.0 ± 29.0

  _____
  Experimental 1S Lambs shift 8159.2 ± 29.0

  Theory $8149.43 \pm .08$

As shown by this table, corrections must be made for a number of systematic shifts of the data, and much of the effort in this experiment was spent in determining these shifts. For the Balmer-β transition the significant corrections are the lineshifts caused by pressure and electric field perturbations in the discharge, and the AC Stark effect, or light shift, due to the polarizing laser beam. For the 1S-2S transition there is a small correction for the AC Stark effect, and a larger correction, the uncertainty of which is the main limitation in this experiment, to compensate for the

frequency shifts introduced by the pulsed amplifiers. This shift is measured by using an interferometer to obtain simultaneous spectra of the cw beam and the pulsed output, but assymetry and "chirping" in the pulsed laser spectrum causes substantial uncertainty in this correction.

For the ground state Lamb shifts we have obtained 8149. ± 29 MHz in hydrogen and 8182.6 ± 29 MHz for deuterium, with similar systematic corrections. These numbers are in excellent agreement with the theoretical results of 8149.43 ± .08 MHz and 8172.23 ± .12 MHz [5].

Because the two-photon lines are narrower and less noisy than the previous measurement [4] it was possible to also make a substantially improved measurement of the Lyman-α isotope shift between hydrogen and deuterium. The result was 670992.3 ± 6.3 MHz which agrees well with the theoretical value of 670994.96 ± .81 [5] and gives the first experimental confirmation of the 11.9 MHz correction due to the recoil of the nucleus [8].

The possibilities for further dramatic improvements in high resolution laser spectroscopy of hydrogen are by no means exhausted. The 1S-2S two-photon transition has a natural line width of only 1 Hz, and attempts to improve the spectral resolution by cw laser spectroscopy of an atomic beam, or by optical Ramsey-fringe spectroscopy with separate oscillatory fields are presently underway [9]. Only a twentyfold improvement in the accuracy of the H-D isotope shift is needed to obtain a new precise value of the electron/proton mass ratio. Simultaneous observation of the Balmer-β line in a beam of metastable hydrogen atoms promises important future improvements in the accuracy of the ground state Lamb shift, and may well turn such a measurement into one of the most stringent present tests of QED calculations.

1. W. E. Lamb and R. C. Retherford, Phys. Rev. 72, 241 (1947); Phys. Rev. 79, 549 (1950).
2. J. Brodsky and S. Drell, Annual Rev. of Nuclear Sci., 20, 147 (1970).
3. T. W. Hänsch, S. A. Lee, R. Wallenstein, and C. Wieman, Phys. Rev. Letters 34, 307 (1975).
4. S. A. Lee. R. Wallenstein, and T. W. Hänsch, Phys. Rev. Letters 35, 1262 (1975).
5. G. Erickson, private communication.
6. N. Bloembergen and M. Levenson, *High Resolution Laser Spectroscopy* (Springer-Verlag, 1976), pg. 315.
7. C. Wieman and T. W. Hänsch, Phys. Rev. Letters 34, 1170 (1976).
8. H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One and Two Electron Atoms* (Academic Press, New York, 1957), Sec. 42.
9. T. W. Hänsch, Physics Today, 30, 34 (1977).

APPENDIX C

Coherent Two-Photon Excitation by Multiple Light Pulses*

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We have studied Doppler-free two-photon excitation of atoms with a train of phase-coherent standing-wave light pulses, originating from the same laser pulse. Quantum interference effects produce narrow spectral fringes which have a physical origin similar to that of Ramsey fringes. Linewidths much less than the Fourier-transform limit of an individual light pulse have been observed for the sodium 3S-5S transition; and a dramatic enhancement of the resonant signal is possible.

We have studied Doppler-free two-photon excitation of atoms with a train of phase-coherent standing-wave light pulses, produced by multiple reflections of a single laser pulse inside an optical resonator. Quantum interference effects result in narrow spectral lines, whose widths depend only on the resonator losses and on the natural transition linewidth, not on the laser linewidth. The resonant excitation probability is proportional to the square of the number of pulses incident during the atomic relaxation time; and a dramatic signal enhancement over single-pulse excitation is possible. We have observed these coherence effects for the sodium 3S-5S transition.

The physical origin of the narrow interference fringes is similar to that of Ramsey fringes.[1] The light field induces coherent atomic oscillations at the two-photon resonance frequency, and the incremental effect of each pulse depends on the phase of its light field relative to these atomic oscillations. It has recently been pointed out[2] that Doppler-free two-photon excitation with standing light waves should permit the observation of optical Ramsey fringes with two spatially

separated fields interacting with an atomic beam, because the phase of the sum frequency is not affected by the first-order Doppler effect. (The atoms in a standing wave see opposite Doppler shifts for the two traveling-wave components.) Optical Ramsey fringes have very recently been observed with two phase-coherent light pulses, separated in time rather than in space.[3]

The presently proposed technique of multiple-pulse interference can provide sharp lines rather than the sinusoidal pattern of two-field Ramsey fringes. This is of particular interest for spectroscopic applications, because it makes it possible to resolve closely spaced line components. Also the large possible signal enhancement is advantageous for the study of weak transitions or for excitation with ultraviolet light where powerful pulsed lasers often with large linewidths are required for the efficient generation of the tunable radiation by nonlinear frequency mixing. The signal enhancement should also permit the use of larger, less intense beams, thus alleviating the problems of ac Stark shifts[4] and transit-time broadening. The large enhancement can even make two-photon excitation attractive for effective pumping of excited states for such applications as selective photochemistry.

To describe the excitation process, we refer to the experimental configuration shown in Fig. 1. A laser pulse, tuned to excite a two-photon transition between atomic levels $|g\rangle$ and $|f\rangle$, enters a resonator of length L, for example, through a partially transmitting mirror, and bounces back and forth, producing a train of phase-coherent pulses, separated in time by $\tau = 2L/c$. The atoms are placed near one mirror, where the incident and reflected fields form a standing wave. The fluorescence from the upper state is monitored as a function of the resonator length.

To show how the multiple pulse excitation leads to narrow linewidths and large signals, we calculate the excitation probability, using perturbation theory. Since we are only interested in the Doppler-free part, we ignore the velocity dependence

FIG. 1. Experimental arrangement.

of the atoms. (The motion of the atoms does not cause dephasing since the atoms see opposite phase shifts for the two counterpropagating waves in the standing-wave field.)

The field for the train of pulses in the resonator is

$$\vec{E}(t) = \sum_{n=0}^{\infty} \vec{E}_0(t - n\tau) \exp[-i\omega(t - n\tau)](R_1 R_2)^{n/2} + \text{c.c.}, \qquad (1)$$

where τ is the roundtrip time in the resonator, R_1 and R_2 are the mirror reflectivities, ω is the laser frequency, and $\vec{E}_0(t)$ is the (slowly varying) envelope function for a single laser pulse inside the resonator. We note that the pulse does not have to be transform-limited. The detailed spatial field distribution is unimportant in the limit of small excitation probabilities, as long as only the fundamental TEM_{00} cavity modes are excited. The interaction Hamiltonian $-e\vec{r}\cdot\vec{E}$ can be written as $V\exp(-i\omega t) + V^*\exp(i\omega t)$, where V is a slowly varying function of time.

As long as the laser frequency is tuned away from any intermediate single-photon resonance frequency ω_{mg}, first-order perturbation theory yields only rapidly oscillating state amplitudes. But second-order perturbation theory[5] predicts a slowly varying amplitude C_f of the final state, describing two-photon excitation. Neglecting atomic relaxation, we expect

$$C_f^{(2)}(t) = \int_0^t dt' \sum_m \frac{i}{\hbar^2} \frac{V_{fm} V_{mg} \exp[i(\omega_{fg} - 2\omega)t']}{\omega_{mg} - \omega}. \qquad (2)$$

If the successive light pulses do not overlap in time, the integral of Eq. (2) can be replaced by the sum of integrals over individual pulse contributions. Since the successive pulses are replicas of each other, except for a constant amplitude factor, only one such integral needs to be evaluated, corresponding to the state amplitude $C_f(\tau)$ after the first pulse. The total sum is then

$$C_f(\infty) = C_f(\tau) \sum_{n=0}^{\infty} \{e^{i\omega_{fg} n\tau}(R_1 R_2)^n\}. \qquad (3)$$

The entire dependence on the laser frequency is contained in $C_f(\tau)$. The sum over the amplitude and phase factors depends only on the atomic resonance frequency ω_{fg} and on the resonator properties, not on the laser frequency ω. Similar summations are encountered in the theory of optical multiple-beam interference,[6] and it is hence not surprising that the excitation $|C_f(\infty)|^2$ versus delay time τ exhibits many of the features of a Fa-

ory-Perot interferogram.
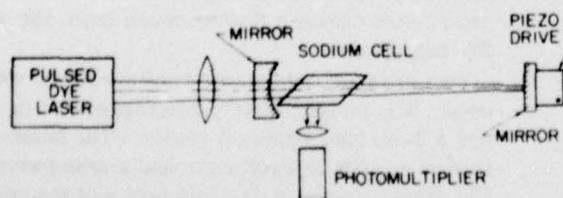
It is not difficult to include in this calculation a (transverse) relaxation rate γ for the oscillating atoms. The total fluorescence from this state $|f\rangle$, excited by one pulse train, is then $P = P_0 \eta$, where P_0 is the fluorescence signal produced by a single pulse, and the factor η describes the multipulse interference:

$$\eta = \eta_{max}\left[1 + F \sin^2(4\pi L / \lambda_0)\right]^{-1},$$

$$\eta_{max} = \frac{(1 + R_1 R_2 e^{-\gamma\tau})}{(1 - R_1^2 R_2^2)(1 - R_1 R_2 e^{-\gamma\tau})},$$ (4)

$$F = 4 R_1 R_2 e^{-\gamma\tau}/(1 - R_1 R_2 e^{-\gamma\tau})^2.$$

Here, $\lambda_0 = 2(2\pi c / \omega_{fg})$ is the resonance wavelength of the two-photon transition. The dependence of the signal on the exact resonator length, described by Eq. (4), is similar to the transmission of a Fabry-Perot interferometer, given by the Airy formula of Ref. 6, except that the resonant maxima occur twice as frequently, and the finesse in the limit of zero atomic linewidth drops with $R_1 R_2$, rather than with $(R_1 R_2)^{1/2}$. The linewidth (full width at half-maximum) of an individual fringe is approximately

$$(\delta\nu)_{FWHM} = (2/\pi\sqrt{F})(c/4L).$$ (5)

The resonant signal enhancement, in the limit of negligible atomic relaxation, is given by $\eta_{max} \approx 1/(1 - R_1 R_2)^2$, which is about equal to the square of the effective number of pulse roundtrips, N^2. In addition, there will be a Doppler-broadened background due to traveling-wave excitation,[7] which reduces the fringe contrast. But this background grows only linearly with N so that the contrast improves with increasing N.

While the coherence aspects of two-photon excitation with multiple pulses can best be understood by working in the time domain, the spectroscopic aspects are clearest in the frequency domain. In some ways the situation resembles two-photon spectroscopy with a multimode laser.[8] Figure 2 shows the intensity spectra of a train of Gaussian light pulses for two different cavity tunings. Resonant two-photon excitation is possible whenever the frequencies of any two peaks add up to the atomic transition frequency ω_{fg}. As illustrated in Fig. 2, this can occur in two ways: Either a peak coincides with $\frac{1}{2}\omega_{fg}$ (solid curve), or the frequency $\frac{1}{2}\omega_{fg}$ falls exactly halfway between two cavity modes (dashed curve). In either case, all the modes under the single-pulse envelope contribute to the excitation. For long pulses with narrow spectral envelope, the excita-
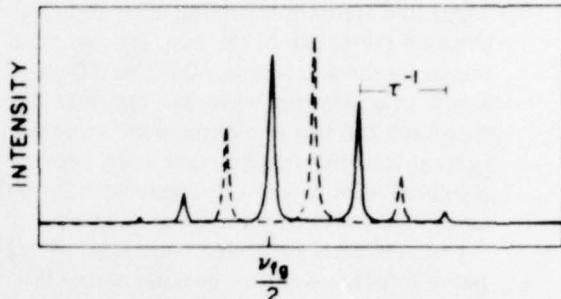

FIG. 2. Intensity spectra of a damped train of Gaussian pulses for a resonator with $R_1 R_2 \approx 0.7$. Dashed and solid curves are for two different resonator tunings. The slowly varying envelope corresponds to the spectrum of one individual pulse.

tion in the latter case may be smaller than in the first case.

To demonstrate multipulse two-photon excitation in atomic sodium, we used the experimental configuration shown in Fig. 1. Short laser pulses were injected into an optical resonator, containing a sodium cell near one end mirror, to provide a standing-wave field. A nitrogen-laser–pumped dye-laser system[9] was used to produce 10-kW pulses of linearly polarized light at about 6022 Å. The pulses were 6 nsec long and the bandwidth was slightly greater than the Fourier-transform limit of about 170 MHz. The resonator was mounted on a rigid optical bench. Its length was maintained by quartz tubes and Invar rods, which provided mechanical and thermal stability, and an insulating box reduced thermal drifts to a few megacycles per minute. One end mirror was mounted on a piezotranslator, permitting continuous scanning over several wavelengths.

The sodium was contained in an evacuated quartz cell with high-quality Brewster windows in order to minimize losses in the resonator. The cell was heated in an electrical oven to about 120 C, producing a sodium density of 2×10^{16} atoms/cm³. The laser was tuned to one hyperfine-structure component of the 3S-5S transition,[7] with an upper-state lifetime of 71 nsec[10]; and the excitation was monitored by observing the ultraviolet cascade fluorescence from the 4P-3S transition.

Two different resonator configurations were used: a 1-m-long nearly hemispherical cavity and a 2-m-long confocal cavity. The beam-entrance mirror in each case had a transmission of 7%. Light losses at the mirrors and the windows resulted in an effective loss factor $R_1 R_2$ of about
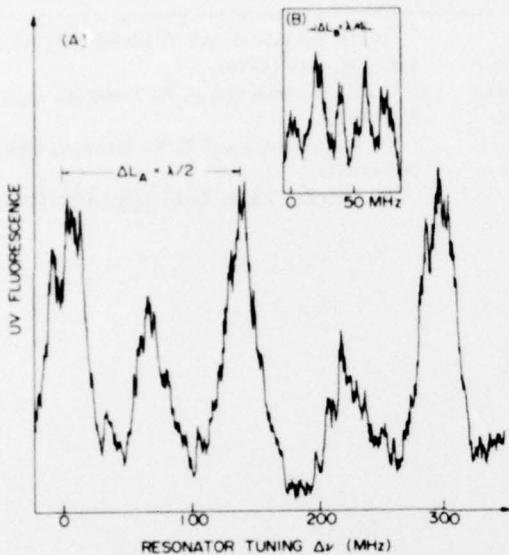
FIG. 3. Multipulse two-photon excitation of the $F = 2$ hyperfine component of sodium 3S-5S. The fluorescence is recorded versus resonator tuning for (a) a 1-m nearly hemispherical resonator and (b) a 2-m confocal resonator.

0.8, reducing the effective number of roundtrips to about 6. The beam waist at the sodium cell was about 1 mm. For the intensities inside the resonator, the resulting ac Stark shifts are only a few megacycles.[4]

The fluorescence versus resonator tuning for the 1-m hemispherical cavity is shown in Fig. 3(a). The spectrum was recorded at a 15-pps laser repetition rate with a 3-sec time constant during about 150 sec. Since the transverse modes of this resonator are not degenerate, it was essential to mode-match in order to excite predominantly the TEM_{00} mode. The observed linewidth is about 30 MHz, significantly less than the laser bandwidth, but larger than predicted by Eq. (5), largely because of residual excitation of higher-order transverse modes. As predicted, the peaks are separated by $\Delta L = \frac{1}{4}\lambda_0$. Since the laser bandwidth is comparable to the resonator free spectral range, the two different resonance conditions illustrated in Fig. 2, produce differently sized peaks, and result in the observed alternation of intensities. Figure 3(b) shows a corresponding spectrum recorded with the 2-m confocal resonator which does not require mode matching. Here the linewidths are only about 12 MHz, presumably limited by cavity vibrations and ac Stark shifts, but the peaks are separated by only $\Delta L = \frac{1}{8}\lambda_0$, since both even and odd resonator modes were ex-

cited.

The interpretation of such a spectrum with its multiple orders is similar to that of a conventional Fabry-Perot spectrum. If two closely spaced lines are present, two combs of resonances appear, separated by the atomic line splitting modulo $c/4L$. In our preliminary experiments, the frequency of the pulsed dye laser was left fixed, and only the external cavity was scanned. For a number of experiments envisioned, it would seem desirable to tune the laser as well. If its frequency were locked to the external resonator, the central fringe of the multipulse interference pattern would always coincide with the atomic transition frequency, as in the double-pulse Ramsey-fringe experiment.[3] One can derive this simply using Eq. (4) by adding the constraint that $L = \frac{1}{2} n \lambda_{\text{laser}}$, where n is integral.

In the present experiment, much laser light is lost when the pulse enters the resonator through one end mirror. In the absence of absorption losses, the input transmittance is $1 - R_1$; and since the excitation is proportional to the intensity squared, the excitation by a single pulse would be decreased by a factor of $(1-R_1)^2$. However, the observed resonant fluorescence in our experiment was actually comparable to the signal produced by a single unattenuated pulse outside the resonator, giving evidence for the predicted enhancement. Current electro-optic technology makes it entirely feasible to inject a laser pulse into a resonator without attenuation. A low-loss cavity, which permits about 100 pulse roundtrips, would then allow an actual enhancement of the excitation probability by a factor of 10 000.

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[1]N. F. Ramsey, *Molecular Beams* (Oxford Univ. Press, London, England, 1956), p. 124.

[2]Ye. V. Baklanov, V. P. Chebotayev, and B. Ta. Dubetsky, Appl. Phys. 11, 201 (1976).

[3]M. Salour, Bull. Am. Phys. Soc. 21, 1245 (1976); M. M. Salour and C. Cohen-Tannoudji, this issue [Phys. Rev. Lett. 38, 757 (1977)]. [Note added.—Optical Ramsey fringes have also been frequently observed in saturated absorption with spatially separated fields; J. C. Bergquist, S. A. Lee, and J. L. Hall, Phys. Rev. Lett. 38, 159 (1977)].

[4]P. F. Liao and J. E. Bjorkholm, Phys. Rev. Lett. 33,

128 (1974).

[5]A. Gold, in *Quantum Optics, Proceedings of the International School of Physics, "Enrico Fermi," Course XLII,* edited by R. J. Glauber (Academic Press, New York, 1969), p. 397.

[6]M. Born and F. Wolf, *Principles of Optics* (Pergamon, Oxford, England, 1970), p. 323.

[7]M. D. Levenson and N. Bloembergen, Phys. Rev. Lett. 32, 645 (1974).

[8]D. E. Roberts and E. N. Fortson, Opt. Commun. 14, 332 (1975).

[9]R. Wallenstein and T. W. Hänsch, Opt. Commun. 14, 353 (1975).

[10]D. Kaiser, Phys. Lett. 51A, 375 (1975).

APPENDIX D

MULTIPLE COHERENT INTERACTIONS

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The observation of narrow optical Ramsey fringes in two-photon excitation of atoms with separate short light pulses, as discussed by M. SALOUR in a preceeding paper, can greatly enhance the potential of Doppler-free laser spectroscopy. It can, in particular, extend the accessible wavelength range into the ultraviolet and vacuum ultraviolet, where there are presently only pulsed tunable lasers available which have by necessity relatively large bandwidths.

At Stanford we began about one year ago to explore the merits of coherent two-photon excitation with more than two light pulses (1), and it soon became apparent that such multi-pulse spectroscopy can offer important advantages in resolution and signal strength over two-pulse experiments.

To better understand the principles and advantages of such an approach, let us briefly recall the origin of two-pulse Ramsey fringes. As illustrated in Fig. 1, top, a single short laser pulse can, at best, produce a broad spectrum whose transform-limited width is about equal to the inverse pulse length. Two-photon excitation with two identical separate pulses produces a sinusoidal spectral interference pattern, as shown at the center of Fig. 1. The first pulse leaves the atoms in a coherent superposition of upper and lower states, and causes atomic oscillations at the two-photon resonance frequency. The effect of the second pulse depends then on the relative phase between these atomic oscillations and the light field. The atoms are either further excited, or they can return to the ground state by stimulated two-photon emission. If the pulse delay is varied in proportion to the laser wavelength, the spacing between the resulting fringes (in frequency) is equal to the inverese delay time between the pulses, while the envelope corresponds to the spectrum of a single pulse.

The width of these two-pulse Ramsey fringes can in principle be arbitrarily narrow, down to the natural atomic linewidth, if only the two light pulses are sufficiently far apart in time. But the number of fringes within the pulse bandwidth increases accordingly, and can easily become confusingly large. More seriously, the sinusoidal fringe pattern makes it difficult, if not impossible, to resolve closely spaced line components, even if the number of fringes remains small.

It is easy to recognize a solution to these problems, if we observe the similarity between the spectrum of such a two-pulse excitation experiment and the well known spatial interference pattern that is obtained in the diffraction of light from a double slit. Based on this analogy we should expect that coherent excitation with a whole train of equidistant identical light pulses will produce a spectrum that resembles the interference

pattern from an array of slits or a diffraction grating. As indicated in Fig. 1, bottom, the fringe spacing should remain the same for a given pulse separation, but the fringes should condense into narrow spectral lines, which correspond to the diffraction orders of a grating. The fringe width should now be determined by the length of the entire pulse train, as long as the transverse relaxation of the atomic oscillators remains negligible.

LIGHT FIELD   TWO-PHOTON SPECTRUM
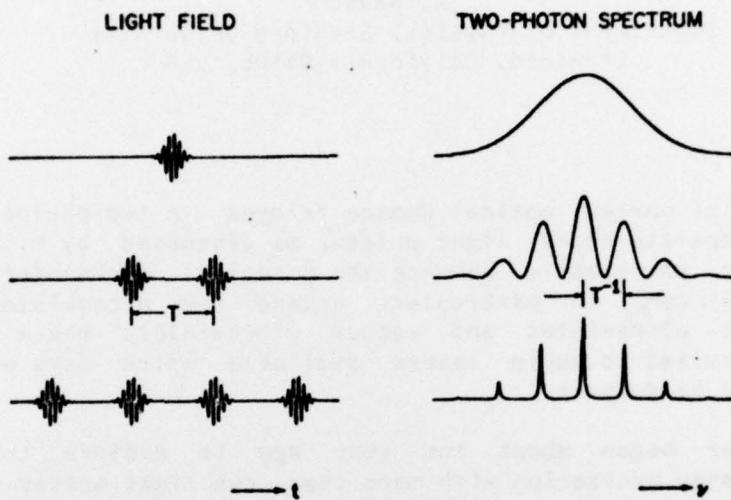
Fig. 1 Short light pulses (left) and resulting Doppler-free two-photon spectra (right). One single pulse produces a broad, transform-limited spectrum (top). Two successive pulses give rise to sinusoidal optical Ramsey fringes (center). A train of multiple identical pulses can produce narrow spectral lines (bottom).

Such multi-pulse excitation is obviously analogous to molecular beam spectroscopy with multiple, spatially separated rf field regions, and it is interesting to note the N. RAMSEY, twenty years ago, has already considered up to four separated oscillatory fields (2). But unlike in this latter case, the complexity and expense of an apparatus for optical two-photon excitation does not have to grow with increasing number of pulses.

The signal magnitude produced by two or more light pulses can be readily calculated by second order perturbation theory, as long as the total two-photon excitation probability remains small (3). Alternatively, we can describe an atom in a coherent superposition of ground state and excited state by a 3-dimensional Feynman vector (4), whose vertical component gives the difference of the level populations or the excitation probability. Its horizontal components give the atomic oscillation amplitude and phase, and can be made to stand still in the absence of light fields by going into a rotating coordinate system. Just as in the familiar case of single-photon excitation of a two-level atom, the two-photon excitation by a light pulse can be described as a finite rotation of this Feynman vector around some external "torque" vector. (This is true even for pulse which is chirping in frequency or otherwise not transform limited, although the net rotation will then be the result of a more complicated detailed angular motion.) In order to predict the excitation by a train of identical light pulses, we

need only look at the corresponding sequence of rotations. The relative angle between subsequent rotation vectors, as seen in the rotating frame, is simply determined by the delay time between the pulses. At exact resonance, all rotations add constructively, and the total excitation probability, to lowest order, is proportional to the square of the number of light pulses in the train. For a given pulse energy, coherent multi-pulse excitation can hence lead to a dramatic signal enhancement. Such an enhancement is often extremely welcome in two-photon spectroscopy, because it can permit one to avoid tight focusing of the laser light with its associated problems of transit time broadening and light shifts.

There are several simple ways of generating a proper train of light pulses experimentally. One attractive possibility is the use of a mode-locked cw dye laser. To control the exact pulse roundtrip time inside the laser resonator, one can employ similar stabilization schemes as are commonly used for single-frequency dye lasers. The accessible wavelength range of such devices is unfortunately still limited, but the possible high pulse peak power can permit an efficient wavelength extension by nonlinear frequency mixing in crystals or gases. We are presently exploring the merits of this approach.

Another, more general scheme, has been employed in a recent experiment by R. TEETS and J. ECKSTEIN in our laboratory (3). Here, the pulse train is generated by injecting a single 7 nsec long light pulse from a nitrogen-pumped dye laser into a simple optical resonator, formed by two mirrors. The light enters the resonator through one of the partially transmitting mirrors. The gas sample is placed near one end mirror, where the atoms see a pulsed standing wave field once during each pulse roundtrip. The roundtrip time is controlled by changing the resonator length with a piezotranslator. As long as the laser bandwidth is large compared to the free spectral range of the resonator, the fringes can be observed by simply keeping the laser frequency fixed, and only tuning the length of the passive resonator. The Na 3s-5s transition was studied in this initial experiment, and the two-photon excitation was observed by monitoring the 3p-3s UV fluorescence light. Fringes of very high contrast and of a few MHz width, much below the 300 MHz laser line width, were easily observed with a 2 m long confocal resonator and about 6 effective pulse roundtrips.

This simple setup immediately suggests an alternative interpretation of the multi-pulse Ramsey fringes. We can ascribe these fringes simply to the discrete axial modes of the passive resonator. This resonator filters narrow spectral lines out of the broadband spectrum of the laser pulse. We could also say that modes within the laser bandwidth are shock-excited by the incoming laser pulse. A fringe maximum is observed if the resonator is tuned so that its modes can excite the two-photon resonance. In this description, multi-pulse two-photon spectroscopy does not seem much different from conventional Fabry-Perot spectroscopy, and one may question whether we are entitled at all to use the term "Ramsey fringes" to describe the observed narrow spectral lines.

It is instructive to look at the two-pulse experiment of M. SALOUR and C. COHEN TANNOUDJI (4) from the same point of view. The optical delay line, which produces the second pulse, can also be considered as a passive optical filter. Similar to a Michelson interferometer, it filters a comb of narrow spectral bands out of the original broad pulse spectrum, and the

observed Ramsey fringes can again be simply ascribed to this spectral filtering. Because of the uncertainty principle, such filtering can, of course, not be performed without changing the pulse shape in time. And the temporal effect of the considered comb-filter is simply the generation of a second, delayed pulse.

Such a description in terms of passive spectral filtering indicates that the exact central frequency of the laser pulse, if it can be defined at all, plays no important role in such experiments. In practice it can nonetheless be highly desirable to employ a cw oscillator with pulsed amplifier and to lock the oscillator frequency to the passive filter or resonator, as in the experiment of SALOUR, if for no other reason than to monitor and control the exact position of the filter transmission bands.

Have we now succeeded in explaining a quantum interference effect without resorting to quantum mechanics? A description simply in terms of passive spectral filtering would certainly seem adequate for weak single-photon excitation of atoms at rest. The Schrödinger equation provides the atoms simply with a means to take the Fourier transform of the incoming pulse train and to analyze the filtered spectrum. For two-photon excitation, however, the situation is not quite as simple. Here, we can satisfy the resonance condition by tuning one of the cavity modes to half the transition frequency. The atom can then be excited by absorbing two photons from this mode. But it can also absorb one photon from the next higher mode and one from the next lower one and so forth. As a consequence, all the modes of the resonator contribute simultaneously to the resonant excitation. The same is also true, if half the transition frequency falls exactly halfway in between two resonator modes. In order to predict the signal magnitude, we hence need to know the relative phases of all oscillating modes. And it is only these phases that imply that, in the experiment of TEETS and ECKSTEIN, for instance, the atomic sample has to be placed near one resonator mirror. The description in the time domain, which seems certainly more intriguing, can then actually be simpler as well.

Nonetheless we can use our spectral filter arguments to explain the predicted strong resonant signal enhancement in multi-pulse excitation. If we send a single light pulse through an optical resonator, consisting of two lossless mirrors of equal reflectivity R, then the filtered, transmitted light (a damped pulse train) will exhibit a comb-like spectrum of narrow lines. The light inside the resonator has obviously the same spectrum, but its intensity is higher by a factor $1/R$, i.e. the spectral energy density in the line maxima is increased over that of the original pulse. But such a passive interferometer still wastes a large portion of the incoming light by reflection. One might say that this reflection simply discards the unwanted light outside the narrow transmission bands of the resonator. But we have to remember that the dominant loss occurs during the initial injection and that the reflected pulse has exactly the same shape as the "good" light that is transmitted into the resonator. It is hence possible to avoid any energy loss by injecting the entire pulse actively into the resonator, for instance with the help of a fast electrooptic or acoustooptic light switch. The signal magnitude can then be many orders of magnitude higher than for single-pulse excitation outside the resonator. The active pulse injection is in fact changing the spectral energy distribution of the available light, and is condensing a broadband spectrum into narrow channels.

Regardless of its interpretation, coherent two-photon spectroscopy with multiple light pulses promises to become a valuable tool for high resolution spectroscopy of atoms and molecules. We expect, for instance, that it should be quite feasible to observe the hydrogen 1S-2S two-photon transition at 2430 Angstroms in this way with a resolution of about 50 kHz, close to the limit set by the transverse relativistic Doppler effect at room temperature. Such a resolution would be more than sufficient for several important new precision measurements. Moreover, the possible strong signal enhancement may make multi-pulse two-photon excitation attractive for the effective excitation of high atomic and molecular levels, for applications such as selective photochemistry.

REFERENCES

1. T. W. Hänsch, in "Tunable Lasers and Applications," A. Mooradian et al., eds., Springer Series in Optical Sciences, Vol. 3, Berlin, Heidelberg, New York 1976, p. 326

2. N. F. Ramsey, Phys. Rev. 109, 822 (1958)

3. R. Teets, J. Eckstein, and T. W. Hänsch, Phys. Rev. Letters 38, 760 (1977)

4. R. P. Feynman, F. L. Vernon, Jr., and R. W. Hellwarth, J. Appl. Phys. 28, 49 (1957)

5. M. M. Salour and C. Cohen-Tannoudji, Phys. Rev. Letters 38, 757 (1977)